# Ultrafast Hole-Spin Dynamics in Optically Excited Bulk GaAs

Michael Krauß and Hans Christian Schneider*
*Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, 67663 Kaiserslautern, Germany*

David J. Hilton†
*Department of Physics, University of Alabama at Birmingham, Birmingham, AL, 35294-1170*

(Dated: December 21, 2010)

We present experimental and theoretical results on hole-spin dynamics in bulk GaAs after ultrafast optical excitation. The experimental differential transmission are compared with a dynamical calculation of the momentum-resolved hole distributions, which includes the carrier-carrier, carrier-phonon and carrier-impurity interaction at the level of Boltzmann scattering integrals. We obtain good agreement with the experimentally determined hole-spin relaxation times, but point out that depending on how the spin-polarization dynamics is extracted, deviations from an exponential decay at short times occur. We also study theoretically the behavior of the spin-relaxation for heavily p-doped GaAs at low temperatures.

Research on spin dynamics and spintronics in semiconductors and metals covers an immense variety of applications in information storage and manipulation in carrier spins in solid state systems.[1–3] Both storage and manipulation of spins are limited by spin relaxation, so that an understanding of spin relaxation processes is important.[4,5] For ultrafast spin dependent dynamics on picosecond timescales, simplified relaxation-time approximations are no longer justified and a fully *microscopic* understanding of the complex spin dynamics is needed.[6–8] While electron spin-dynamics continue to be extensively investigated in semiconductors due to their extremely long spin-relaxation times,[9] hole-spin dynamics are intriguing for different reasons. In III-V semiconductors, hole dynamics is inherently different from that of electrons because of the strong spin-orbit coupling of the hole states. Thus, hole-spin relaxation and momentum (energy) relaxation occur on a comparable ultrashort timescale.

In this paper, we connect differential transmission measurements of the hole-spin dynamics[10] with a microscopic calculation that includes the anisotropic band-structure as well as carrier-carrier and carrier-phonon/impurity scattering mechanisms at the level of Boltzmann scattering integrals. Although our approach does not take into account all the intricacies of the dynamical multi-band screening (including phonon-plasmon coupling)[11,12] and interband polarizations, it captures essential aspects of the spin dynamics of holes.[13] We show that this calculation compares well with measured hole-spin relaxation times, but that deviations from an exponential spin decay occur for short times depending on how the relaxation time is extracted. Further, we present results for the hole-spin relaxation at high p-doping, which underlies a recent treatment of GaAs-based ferromagnetic semiconductors.[14]

Recent theoretical treatments of hole spin (and charge) dynamics in bulk GaAs have focused on the calculation of energy-dependent spin *relaxation-rates* due to phonon/impurity scattering,[15] the investigation of coherent hole (spin) dynamics in GaAs,[16,17] and the influence of Dyakonov-Perel type precession effects due to spin-dependent splittings between hole bands in GaAs quantum wells.[18] Very recently, additional contributions due to "small" band structure effects and the spin-orbit contribution to the hole-phonon interaction for holes in bulk GaAs have also been analyzed.[19]

From an experimental point of view, hole-spin polarizations can be created efficiently using optical orientation techniques.[20] Because of the ultrashort lifetimes[21] involved, it is difficult to *unambiguously* study hole dynamics in degenerate pump-probe experiments (i.e., experiments with the same pump and probe wavelengths) due to the competing presence of the coherent artifact on the same timescale[22] and, to a lesser degree, the electron spin dynamics.[9] Hole spin study, therefore, requires measurement of the spin-dependent carrier dynamics on ultrashort timescales using a time-delayed, mid-infrared probe beam (i.e., a nondegenerate pump-probe experiment), which was realized only much later[10] than similar experiments on electrons.[9]

We focus here on the dynamics of the spin-polarized heavy hole subband. We have performed nondegenerate, polarization-resolved pump-probe spectroscopy at 300 K to measure the hole spin relaxation lifetime in intrinsic GaAs as a function of input pump fluence, $\mathcal{F}$. In this setup, 1 W of average power from an 80 fs titanium:sapphire laser (Spectra Physics Tsunami) operating at an 80 MHz repetition rate is used to pump a custom-designed optical parametric oscillator based on periodically-poled lithium niobate, as described in further detail in ref. 23. A portion of the titanium:sapphire beam ($\leq 300$ mW) is circularly polarized using a quarter wave plate and is used to photoexcite spin-polarized electron-hole pairs in the GaAs sample. The pump beam induces a circular birefringence and results in a time-dependent rotation of the transmitted probe polarization. To isolate the dynamics of the hole spin state from the electron spin, we probe the occupancy of the heavy hole and/or light hole subbands using a synchronized mid-infrared idler pulse ($\lambda = 3200$ nm), which probes the induced transitions from the split off subband into the heavy hole subband. This wavelength directly probes states from the lower split-off hole subband to the heavy hole

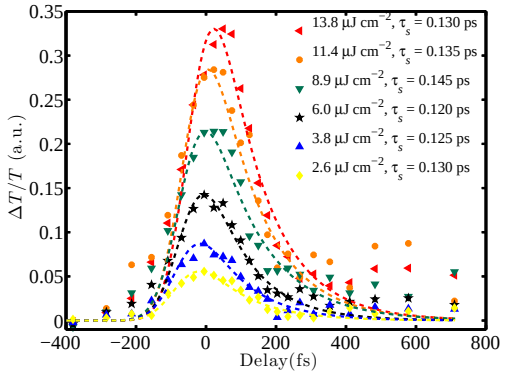


FIG. 1. Fluence dependence of the time-resolved differential transmission, $\Delta T/T(t)$, at 300 K.

subband at the same quasimomentum ($k \approx 0.3$ nm$^{-1}$) as the initial photoexcitation at 800 nm.

Figure 1 shows the measured field rotation, $\Delta T/T(t)$, of the mid-IR probe from $\mathcal{F} = 2.6$ to 13.8 $\mu$J cm$^{-2}$. The input probe beam is linearly polarized ($\hat{x}$), which is an equal admixture of both the co-circularly polarized ($\hat{\sigma}_+$) and counter circularly polarized($\hat{\sigma}_-$). A $MgF_2$ linear polarizer is rotated 90° with respect to the input polarization (i.e., along $\hat{y}$) and is used to measure the field rotation of the transmitted probe, which depends on the time-dynamics of the the heavy hole occupancy state near the quasimomentum, $k$, of photogeneration. Using the measured absorption coefficient of GaAs,[24] the resulting carrier concentrations range from $5\times10^{16}$ cm$^{-3}$ ($\mathcal{F} = 2.6$ $\mu$J cm$^{-2}$) to $2\times10^{17}$ cm$^{-3}$ ($\mathcal{F} = 13.8$ $\mu$J cm$^{-2}$). We fit the measured field rotation to a single exponential model and account for the finite pump and probe pulse widths by convolving the model function with a gaussian with a width of 120 fs (see ref. 10 for more details of the fitting model). We find no significant trend in the extracted lifetimes, which range from 120 fs to 145 fs, within the experimental error (±20%) of this experiment.

The calculation of the hole-spin dynamics follow the procedure outlined in ref. 13. The electron and hole states around the fundamental bandgap needed as input for the dynamical calculation are determined from an 8-band Kane model $\mathcal{H}(\vec{k})$ with six hole and two electron bands containing terms up to second order in $k$.[25] By diagonalizing $\mathcal{H}(\vec{k})$ we obtain the "intelligent basis" (in the sense of ref. 26) for the hole states: $|\nu,\vec{k}\rangle$ and their energy eigenvalues $\varepsilon_{\nu,\vec{k}}$. The label $\nu = (b,p)$ includes the band index $b$ = E, HH, LH, SOH, (for electrons, heavy holes, light holes, and split-off holes) and the pseudospin $p = 1, 2$. The pseudospin can be introduced because the quasiparticle dispersions of all four types of carriers are (nearly) doubly degenerate.

The dynamical equations governing the time evolution of the incoherent carrier distributions, $n_{\nu,\vec{k}}$, under the influence of optical fields and scattering are:[13,27]

$$\frac{\partial}{\partial t} n_{\nu,\vec{k}} = \Gamma^{\rm in}_{\nu,\vec{k}}(1 - n_{\nu,\vec{k}}) - \Gamma^{\rm out}_{\nu,\vec{k}} n_{\nu,\vec{k}} \tag{1}$$

(plus a contribution from the optical excitation, which neglects the hole band coherences between hole states of the "intelligent basis," or, equivalently, only describes contributions to the hole spin relaxation of the Elliott type[26] that arise from the $\vec{k}$ dependent mixing of different spins in the states $|\nu,\vec{k}\rangle$. Contributions to the spin relaxation via coherent spin precession,i.e., Dyakonov-Perel-type spin dynamics,[19] are neglected.

The dynamical in-scattering rate consists of the carrier-carrier interaction contribution (i.e., eq. (2) in Ref. 13) and the carrier-phonon interaction

$$\Gamma^{\rm in}_{\nu,\vec{k}}|_{\rm c-p} = \frac{2\pi}{\hbar} \sum_{\nu_1,\vec{q},\lambda} |M_{q,\lambda}|^2 |\langle \nu_1, \vec{k}+\vec{q}|\nu,\vec{k}\rangle|^2 n_{\nu_1,\vec{k}+\vec{q}} [(1+N_{q,\lambda})\delta(\Delta E_-) + N_{q,\lambda}\delta(\Delta E_+)] \ . \tag{2}$$

where $\Delta E_\pm = \varepsilon_{\nu,k} - \varepsilon_{\nu_1,|\vec{k}+\vec{q}|} \pm \hbar\omega_{q,\lambda}$. There is also a contribution similar to eq. (2) due to carrier-impurity scattering.[7] Here, $N_{q,\lambda}$, $M_{q,\lambda}$, and $\omega_{q,\lambda}$ are the phonon occupation numbers, carrier-phonon coupling matrix elements, and phonon dispersions for LO and LA phonons, respectively.[28] The out-scattering rates $\Gamma^{\rm out}$ are obtained from $\Gamma^{\rm in}$ by exchanging $(1-n)$ with $n$ and $(1+N)$ with $N$. Equation (2) and its counterpart for Coulomb scattering describes two-particle scattering processes connecting states $|\nu,\vec{k}\rangle \to |\nu_1,\vec{k}+\vec{q}\rangle$ (and $|\nu_2,\vec{k}_1+\vec{q}\rangle \to |\nu_3,\vec{k}_1\rangle$) with different average spin. The pronounced anisotropy of the single-particle states, which is due to the spin-orbit interaction, is included in our calculation in the overlaps and the carrier energies, $\epsilon_{\nu,\vec{k}}$, which enter the energy-conserving delta functions in the

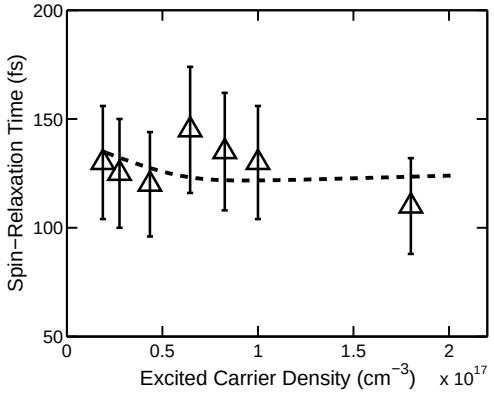


FIG. 2. Theoretical (curve) and experimental values (symbols) for fluence dependence of hole-spin relaxation in intrinsic GaAs at room temperature.

scattering rates. The anisotropies and high-dimensional scattering integrals result in a challenging numerical problem. We incorporate the effects of the anisotropy by expanding $n_{\nu,\vec{k}}(t)$ into spherical harmonics $Y_{\ell,m}(\hat{k})$ up to order $\ell = 4$ and retain only the expansion coefficients with radial or cubic symmetry, as these are the dominant symmetries of the Hamiltonian $\mathcal{H}$.[25] This procedure, together with the use of static screening, $v^s(q) = e^2/[\epsilon_0\epsilon_{bg}(q^2+\kappa^2)]$, reduces the numerical complexity. Additional simplifications are achieved by including only the heavy-hole bands in the dynamical calculation as the number of optically excited light holes is much smaller,[13] and by replacing the non-equilibrium electron distributions by equilibrium distributions with the lattice temperature. We then calculate the spin polarization of the HH band. In ref. 13, it was shown that the spin polarization is not identical to the DT signal that is measured, but the deviations are small enough for the accuracy of the present comparison.

We include the carrier excitation by an ultrashort optical pulse using a $\hat{\sigma}_+$ polarized plane wave traveling in the (001) direction ($\hat{z}$) to generate an initial condition for the carrier distributions

$$n_{\nu,\vec{k}}(t=0) = \sum_{\mu} |\vec{d}_{\mu\nu}(\vec{k}) \cdot \vec{E}|^2 g(\hbar\omega - \varepsilon_{\mu,\vec{k}} - \varepsilon_{\nu,\vec{k}}) \ . \tag{3}$$

Here, $\vec{d}_{\mu\nu}(\vec{k}) = e\langle\mu,\vec{k}|\vec{r}|\nu,\vec{k}\rangle$ are the dipole-matrix elements, $\vec{E}$ is the laser field, and $\hbar\omega$ is the photon energy of the exciting field. A Gaussian broadening function $g(\varepsilon)$ peaked at $\varepsilon = (\hbar\omega - \varepsilon_{\mu,\vec{k}} - \varepsilon_{\nu,\vec{k}})$ accounts for the spectral width of the pulse (15 meV).

To compare with experimental results in Fig. 2, we convert the experimental laser fluence into excited carrier densities, and extract the relaxation time from an exponential fit to the spin polarization at the probe-laser wavelength. The experimental results are in good agreement with our calculations and a recent theoretical study.[19]. Fig. 2 shows that hole spin-relaxation is rather insensitive to excited carrier densities on the order of $10^{17}\,\mathrm{cm}^{-3}$. The calculation also predicts a very weak temperature dependence with a hole spin relaxation time of 250 fs for an excited density of $10^{17}\,\mathrm{cm}^{-3}$ at 4 K, which is somewhat shorter than the result predicted by the Elliott-Yafet spin *relaxation-time*.[15]

In Fig. 3, we simulate the momentum dependence of the spin polarization dynamics. We model the excitation by an 800 nm pump pulse that excites a carrier density of $10^{17}\,\mathrm{cm}^{-3}$. This leads to anisotropic initial hole distributions that are peaked at momentum $k = 0.3\,\mathrm{nm}^{-1}$ (after integration over the angular variables). Fig. 3 shows different spin polarizations calculated using the angle-averaged distributions $n(k,t)$ for different modulus of the hole momentum $k$. The decay of the spin polarization near the peak position of the initial distribution is nearly exponential, as can be seen from the linear curve shape on the logarithmic scale. The evolution of the spin polarization for larger and smaller momenta (i.e, away from the momenta of the initially photoexcited holes) is significantly faster and non-exponential for the initial $\sim 100$ fs. After the initial non-exponential dynamics the spin-relaxation becomes exponential again with the same time constant. The spin relaxation-time therefore depends on the way it is extracted from the calculation, as opposed to the case of electron spin-relaxation in p-doped GaAs where the spin relaxation time can be quite rigorously defined.[29] The dashed lines in Fig. 3 correspond to experimental situations where pump and probe pulses are detuned, as was investigated in ref. 10 (see Fig. 5 therein), where no change in spin relaxation times for different detuning between pump and probe pulses was found. The apparent discrepancy likely results from the analysis of the experimental data via a convolution of Gaussians, which are used to describe both the pulse shapes and the dynamics of the material response. The analysis of the pump-probe experiment therefore presupposed an exponential decay of the polarization, which turns out to be only somewhat longer than the experimental time resolution of about 100 fs[10].

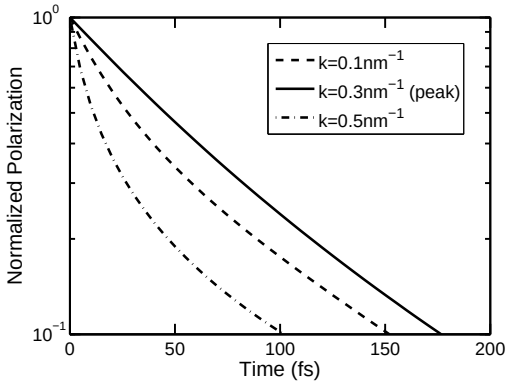


FIG. 3. Computed hole-spin polarization dynamics at different hole momenta for fixed excitation at $k = 0.3\,\mathrm{nm}^{-1}$ in intrinsic bulk GaAs. The excited carrier density is $n = 10^{17}\,\mathrm{cm}^{-3}$.

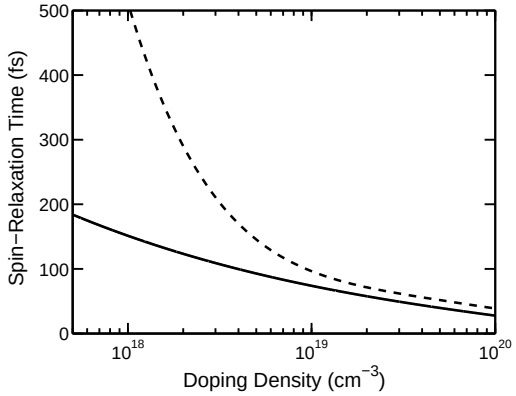


FIG. 4. Computed p-doping dependence of hole-spin relaxation in GaAs at low temperatures (solid line). The dashed line results if only scattering by from ionized donors is included.

For degenerate pump and probe, this analysis yields the exponential decay of the spin polarization in agreement with the calculated result in Fig. 3. For non-degenerate situations the non-exponential features in Fig. 3 on timescales below 100 fs are not resolved. Their resolution would require a time resolution well below 100 fs.

Finally, we examine the hole-spin relaxation dynamics in heavily p-doped GaAs at low temperatures (5 K) theoretically. By choosing these parameters, we estimate the hole-spin relaxation time for GaMnAs in the ferromagnetic phase at low temperatures, which has similar carrier doping levels. We model the optical excitation of nonequilibrium carriers in p-doped GaAs by a pump laser with a wavelength of 400 nm. Choosing this wavelength ensures that the optical transitions are not blocked by holes even for very high doping densities. We assume initial carrier distributions of the form $n_k^0 = n_k^{\mathrm{dop}} + n_k^{\mathrm{exc}}$, which contain both the influence of the doping and the optical excitation. All itinerant holes introduced by the p-doping are assumed to be thermalized at the lattice temperature; they are modeled by unpolarized Fermi-Dirac electron distributions $n^{\mathrm{dop}} = f(\epsilon_k^{\mathrm{e}})$ at lattice temperature with carrier density equal to the density of dopants. For the optical excitation we assume again an ultrashort pulse with a width of 15 meV. Fig. 4 shows that the hole-spin relaxation drops to approximately 10 fs for doping densities typical for GaAs based magnetic semiconductors. At these high doping densities, the rapid momentum relaxation due to scattering from the ionized donors provides the main relaxation mechanism. The assumption of a hole spin relaxation time of ∼10 fs in ref. 14 makes it possible to neglect a spin-bottleneck effect for ferromagnetic GaMnAs. Our results, based on a dynamical calculation, support this assumption.

In conclusion we examined ultrafast hole-spin relaxation using differential transmission measurements and a microscopic theoretical approach for different excitation conditions. Theory and experiment show that the hole-spin relaxation time is approximately 100 fs and independent of moderate changes of the pump fluence at room temperature. The concept of spin-relaxation time is not, in general, applicable for non-degenerate pump-probe schemes, but the deviations from the exponential decay of the spin-polarization are confined to time scales of less than 100 fs, and can therefore only be resolved by a time resolution well below 100 fs. These results also demonstrate the limited validity of a single spin *relaxation-time* for holes. Strong p-type doping shortens relaxation times significantly due to the scattering from ionized donors. The calculated spin relaxation times support the assumption of very fast hole-spin relaxation in ferromagnetic GaMnAs at low temperatures.

We acknowledge support by the DFG through GRK 792 and a grant for CPU time from the the NIC Jülich. We are grateful to C. L. Tang and M. W. Wu for helpful discussions.

---

* hcsch@physik.uni-kl.de
† dhilton@uab.edu

[1] D. D. Awschalom, D. Loss, and N. Samarth, eds., *Semiconductor Spintronics and Quantum Computation* (Springer, Berlin, 2002).
[2] S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnar, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).
[3] I. Žutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 323 (2004).
[4] L. J. Sham, J. Phys.: Cond. Matt. **5**, A51 (1993).
[5] N. S. Averkiev, L. E. Golub, and M. Willander, J. Phys.: Cond. Matt. **14**, R271 (2002).
[6] M. M. Glazov and E. L. Ivchenko, JETP Lett. **75**, 403 (2002).
[7] M. Q. Weng and M. W. Wu, Phys. Rev. B **68**, 075312 (2003).
[8] H. C. Schneider, J. P. Wüstenberg, O. Andreyev, K. Hiebbner, L. Guo, J. Lange, L. Schreiber, B. Beschoten, M. Bauer, and M. Aeschlimann, Phys. Rev. B **73**, 081302(R) (2006).
[9] J. M. Kikkawa and D. D. Awschalom, Phys. Rev. Lett. **80**, 4313 (1998).
[10] D. J. Hilton and C. L. Tang, Phys. Rev. Lett. **89**, 146601 (2002).
[11] J. H. Collet, Phys. Rev. B **47**, 10279 (1993).
[12] M. Woerner and T. Elsaesser, Phys. Rev. B **51**, 17490 (1995).
[13] M. Krauss, M. Aeschlimann, and H. C. Schneider, Phys. Rev. Lett. **100**, 256601 (2008).
[14] Ł. Cywiński and L. J. Sham, Phys. Rev. B **76**, 045205 (2007).
[15] Z. G. Yu, S. Krishnamurthy, M. van Schilfgaarde, and N. Newman, Phys. Rev. B **71**, 245312 (2005).
[16] A. Dargys, Phys. Rev. B **70**, 125207 (2004).
[17] D. Culcer, C. Lechner, and R. Winkler, Phys. Rev. Lett. **97**, 106601 (2006).
[18] C. Lu, J. L. Cheng, and M. W. Wu, Phys. Rev. B **73**, 125314 (2006).
[19] K. Shen and M. W. Wu, Phys. Rev. B **82**, 115205 (2010).
[20] F. Meier and B. P. Zakharchenya, eds., *Optical Orientation* (North-Holland, Amsterdam, 1984).
[21] F. Ganikhanov, K. C. Burr, D. J. Hilton, and C. L. Tang, Phys. Rev. B **60**, 8890 (1999).
[22] C. W. Luo, Y. T. Wang, F. W. Chen, H. C. Shih, and T. Kobayashi, Optics Express, **17**, 11321 (2009).
[23] K. Burr, C. Tang, M. Arbore, and M. Fejer, Opt. Lett. **22**, 1458 (1997).
[24] J. Blakemore, J. Appl. Phys. **53**, 520 (1982).
[25] R. Winkler, *Spin-Orbit Coupling Effects in Two-Dimensional Electron and Hole Systems*, Springer Tracts in Modern Physics, Vol. 191 (Springer, Berlin, 2003).
[26] J. Fabian, A. Matos-Abiague, C. Ertler, P. Stano, and I. Žutić, Acta Physica Slovaca **57**, 565 (2007).
[27] J. H. Collet, Phys. Rev. B **47**, 10279 (1993).
[28] P. Y. Yu and M. Cardona, *Fundamentals of Semiconductors*, 3rd ed. (Springer, Berlin, 2001).
[29] H. C. Schneider and M. Krauß, in *Ultrafast Phenomena in Semiconductors and Nanostructure Materials XIV*, Proceedings of SPIE, Vol. 7600 (SPIE, Bellingham, WA, 2010) p. 1D.